Chapter 4

Experimental setup

4.1 Introduction

In order to measure the tiny recoil momenta of the target particles two conditions have to be fulfilled. Firstly one has to build a spectrometer that precisely images the recoil momentum vector on a detector. Secondly, one needs an extremely cold target, such that the velocity spread of the target atoms is much smaller than the velocity resolution one has to achieve to resolve different channels.

In this chapter an overview of the experimental setup will be given. The cold target is discussed, starting with a brief and general explanation of the principles of laser cooling and trapping, followed by a discussion of the laser, its frequency stabilization and the magneto-optical trap. Thereafter the implementation of the recoil-ion momentum spectroscopy is treated. That discussion includes also the ion source, the design and properties of the recoil-ion momentum spectrometer and the data analysis. Finally, the method to obtain spectra from excited target atoms is dealt with.

The design and construction of the first version of the setup started in 1998. It became operational in the second half of 2000. Many parts of the setup were described earlier in the thesis of Turkstra [147]. Here, a complete overview of the setup will be given, with emphasis on the changes and progress made during this thesis research period.

4.2 Laser cooling and trapping of Na target atoms

Starting off from the principles of laser cooling and trapping and magneto-optical trapping, a detailed description of the experimental implementation of our magneto-optical trap for Na is given, together with some specific properties relevant to the MOTRIMS technique.

4.2.1 Principles of Magneto-Optical Trapping

Laser cooling and trapping makes use of the fact that light carries momentum and that atoms can be deflected by resonant light. Consider a two level atom interacting with a laser beam:
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Figure 4.1: Light forces on a Na atom as a function of its velocity, calculated for two counterpropagating laser beams (solid line) and the individual laser beams (dashed line). The laser frequency is one linewidth red-detuned from the resonance frequency. The laser power in each beam is 20 mW/cm$^2$. Note that the acceleration is $\sim 50,000$ times the gravitational one ($\approx 10$ m/s$^2$).

The atom can be excited from the ground to the excited state by absorption of laser light. Each absorbed photon transfers a small amount of momentum to the atom in the laser beam direction. Because of the finite lifetime of the excited state the atom will decay to the ground state by emitting a photon. Also the emitted photon delivers a momentum kick to the atom. Because the direction in which the photons are emitted is random, the net momentum of the emitted photons cancels after many cycles of absorption and emission. Thus the net force exerted on the atom is in the direction of the laser beam. The Doppler shift associated with the atom’s motion makes this force dependent on the velocity of the atoms. An atom moving against the direction of the laser beam is on resonance if the laser is tuned at a frequency below the atom’s resonance frequency (red-detuned).

In figure 4.1 a typical acceleration curve of an atom between two counterpropagating red-detuned laser beams is depicted. The acceleration (solid curve) is opposite to the atom’s motion (i.e. the atoms are decelerated) but decreases in magnitude as the atom’s velocity becomes too high. The deceleration can exceed $5 \times 10^5$ m/s$^2$. The force of two red-detuned counterpropagating laser beams results in a one dimensional damping of an atom’s motion and is known as optical molasses, i.e., “an atom moving through syrup” [148].

Using three pairs of orthogonal laser beams one can cool the motion of an atom in all directions but one cannot trap this atom, yet. The reason is that the effective laser force approaches zero when the atom’s velocity goes to zero and the atom can effectively creep out of the laser beam. To create a real trap, the light forces must also depend on the atom’s position within the trap. This can be achieved by adding a magnetic field in anti-Helmholz configuration. The magnetic field of such an arrangement is zero in the center but increases
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Figure 4.2: The scheme for a magneto-optical trap as introduced by Raab et al. [20] and used in this work. Six red-detuned circular polarized laser beams are arranged along all six spatial directions. Two coils carrying equal currents in opposite direction create an anti-Helmholtz magnetic field.

in all directions. Atoms outside the center will find themselves in a magnetic field and the Zeeman effect will then change the resonance frequency of the atomic transition because the ground and excited states are affected differently by the magnetic field. The Zeeman effect in conjunction with proper circular polarization of the laser beams (see figure 4.2) causes the atoms to be in resonance only with those laser beams which are directed in opposite direction to the motion of the atoms, using . Thus atoms are pushed back to the center again. The inhomogeneous magnetic field together with the six red-detuned laser beams form the magneto-optical trap (MOT).

Na atoms serve as target in the ion-atom collisions studied in this thesis. Na is one of the most common atomic species in laser cooling and trapping. Because $^{23}\text{Na}$ has a nuclear spin of $I = 3/2$ the electronic levels are hyperfine split. The energy differences between the hyperfine levels of the Na $3s \, ^2S_{1/2}$ ground state and the $3p \, ^2P_{3/2}$ excited state are shown in figure 4.3. Our MOT operates at the $F_g = 2 \rightarrow F_e = 3$ transition frequency, but due to the line width of the transition and the small Doppler and Zeeman shifts present in the MOT, some atoms can be excited accidently to the $F_e = 2$ state and this excited state can also radiate to the $F_g = 1$ ground state, a state not coupled to the laser light. To get these “lost” atoms back into the cooling cycle, a second frequency is used, resonant with the $F_g = 1 \rightarrow F_e = 2$ transition, and is called the “repumping” laser beam. Note that initially the two ground states are populated statistically, i.e., 3/8 in $F_g = 1$. The repumping laser beam also brings these atoms into the cooling cycle.

The first operational MOT dates from 1987 by Raab et al. [20]. In 1990 Monroe et al. [149] demonstrated for the first time a background loaded MOT. The latter type of MOT is the most common nowadays. Laser cooling and trapping has been described extensively in literature and will not be discussed further here. An overview of the development of laser cooling and trapping, the theoretical description, and several experimental methods can be
found in [150, 151]. Details about the principles of laser cooling and trapping of Na within the context of MOTRIMS can be found in [147, 152].

4.2.2 Laser

All the laser beams used in the experiment are provided by a single-frequency dye laser (Model 380D, Spectra-Physics) [153], pumped by a solid state CW laser (Millennia Vs, Spectra-Physics) with an output wavelength of 532 nm and a maximum power of 5 W. In a dye laser the gain medium is an organic dye in a solvent. Dyes are complex molecules with diffuse energy bands, which ensure that the laser is continuously tunable over a broad range (∼ 50 nm). The whole visible wavelength range can be spanned with different dyes, with the restriction that the dyes can only emit photons of lower energy than the ones absorbed. A detailed overview of the working principles of dye lasers can be found in [154].

A schematic of our ring dye laser is shown in figure 4.4. The laser beam in the ring dye laser is folded in a loop by mirrors M₁ to M₄. The pump mirror (M₆PUMP) focuses the pump laser beam into the dye jet. The four mirrors M₁ to M₄ reflect the beam around the cavity, in a bowtie pattern. Mirror M₃ is piezoelectrically driven for active stabilization, the cavity-length is adjusted by a high-voltage piezoelectric translator (PZT) driver.

To scan the cavity mode a dual galvoplate assembly is added. The effective cavity length is changed by counter-rotating two quartz-plates inserted in the cavity at Brewster’s angle. As the plates are rotating the optical path length through the quartz-plates is changed. This means the galvoplates allow smooth scanning of the frequencies without a spatial displacement of
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Figure 4.4: Beam path in the Model 380D cavity. The spherical folding mirrors \( M_1 \), \( M_2 \) and \( M_3 \) and the flat output coupler \( M_4 \) fold the beam in a figure-eight path, which is traversed in the direction shown by arrows. The direction is dictated by a unidirectional device. The dye jet intersects the beam perpendicular. 1. astigmatism compensator, 2. dual galvoplates, 3. unidirectional device, 4. scanning etalon and 5. birefringent filter.

The linewidth of a single-frequency dye laser is determined by the stability of the optical path length of the cavity. Instabilities can be caused by microphonic vibrations, ambient pressure fluctuations and bubbles in the dye jet. Strong perturbations may cause mode-hops, i.e. jumps between cavity modes. In order to achieve the narrowest possible line width and to make the laser virtually immune against mode-hops, the 380D uses a so-called Stabilok System (figure 4.5). The frequency is stabilized by locking it to interference fringes from a Fabry-Perot etalon. Frequency deviations are detected and corrected by error signals fed back to the PZT-mounted \( M_2 \) and the dual galvoplates.

The Stabilok System consists of two interferometers. The reference interferometer, the master, has a free spectral range (FSR) of 0.5 GHz, which means that deviations within a reference fringe can be detected by the master but a mode-hop of only two cavity modes (cavity mode spacing is 200 MHz) would lock the laser frequency to another fringe of the master interferometer. Therefore, a second interferometer, the slave, with a FSR of 10 GHz monitors the laser frequency. When a mode-hop occurs, the slave circuitry takes control and returns the laser frequency to the correct fringe of the master. Then the frequency control is returned to the master circuitry for fine-frequency control.

Both interferometers convert frequency changes to amplitude changes, which are measured by photodiodes. Because the signals arriving at the master and slave depend on both the frequency and the intensity, and only the frequency should be controlled, these signals are normalized to the primary beam that enters the reference station.

Typically the pump laser is set to an output power of 3.5 W, which in the dye laser is converted into 300 to 400 mW of laser light of 589 nm (\( \text{D}_2 \)-line of sodium). The bandwidth of the laser is less than 1 MHz and the frequency drift can be less than 50 MHz/hour. A more detailed description of our laser system can be found in [155].
4.2.3 Repump frequency

An electro-optical modulator (EOM) is used to provide the repump frequency needed to keep all atoms in the cooling cycle. The technique of generating RF sidebands on a laser field is based on the equivalence of phase modulation and frequency modulation. A detailed technical description of the EOM can be found in [156].

During this thesis research two different EOM’s were used, both having a LiTaO$_3$ crystal for sideband generation. The first EOM (home made) operated at 860 MHz. Its first order sidebands are separated by $2 \times 860$ MHz (see figure 4.6). The lowest frequency (-1 sideband) serves as the pump frequency while the highest frequency (+1 sideband) is used for repumping. Two major disadvantages were faced using the 860 MHz EOM. First of all, most of the laser power (30–50\%) resides in the central frequency, which is not used in the experiment. Secondly the pump and repump frequency have the same intensity, which is unnecessary, because a repump intensity of 10–20\% of the pump intensity suffices. Therefore, only effectively 25–35\% of the total laser power is used for cooling and trapping. Because the number of atoms trapped in the MOT, i.e. available for the collision experiment strongly depends on the laser power, a more efficient usage of laser power is highly desirable.

Therefore a new EOM operating at 1720 MHz has been installed (New Focus 4421). Because now the frequency spacing between successive orders is equal to the difference in pump and repump frequency, one can use the main frequency for pumping and the +1 sideband for repumping (see figure 4.6). This solves both problems outlined above. The repump intensity can be tuned and can be set at 10–20\%, leaving 60–80\% in the main frequency for pumping. This increase of effective laser power in the pump frequency resulted in a MOT density increase by a factor of 4.

Figure 4.5: Schematic of the Stabilok System. By a beamsplitter (BS) a small amount of light is taken from the laser beam and fed into the Stabilok System: the incoming beam intensity and the outputs of the master and slave are measured by photodiodes (PD).
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4.2.4 External frequency stabilization

Even though the frequency drift of the laser is less than 50 MHz/h, this should be compensated for in order to be able to run measurements of several hours. The frequency range in which the atoms can be trapped is namely only $\sim 20$ MHz wide. To lock the laser frequency an absolute reference is needed. This signal is provided by means of saturated absorption spectroscopy (figure 4.7). In saturated absorption spectroscopy a strong pump and a weak probe laser beam with equal frequency traverse a Na vapor cell in opposite directions. The absorption of the probe beam is altered by the presence of the pump beam.

If the laser frequency is $\nu_0$, then for atoms moving with a velocity $v$ this frequency is Doppler shifted. For atoms moving antiparallel to the beam direction the frequency is shifted to $\nu_1 = \nu_0 \left(1 + \frac{v}{c}\right)$, while atoms moving parallel see $\nu_2 = \nu_0 \left(1 - \frac{v}{c}\right)$. For certain velocity classes of atoms the frequencies $\nu_1$ and $\nu_2$ are equal to hyperfine transition frequencies. These atoms can absorb photons from the probe beam as well as from the pump beam. Two situations are of importance here:

1. Both beams operate on the same lower level. The pump beam will lower the population of the lower level so there will be less atoms available to absorb photons of the probe beam. This causes a minimum in the probe beam absorption and leads to a maximum in the transmitted probe beam intensity.
2. One of the beams excites atoms in the ground state $F = 1$ and the other one atoms in $F = 2$. The pump beam increases the population of the state on which the probe beam operates, because the state excited by the pump beam can also decay via spontaneous emission to the state on which the probe beam acts. Now there is a maximum in the probe beam absorption, so a minimum in the probe beam intensity.

A chopper regularly interrupts the pump beam. The signal of the photodiode, which measures the intensity of the probe beam, is fed into a lock-in amplifier (FEMTO LIA-MV-150), which subtracts the signals with and without the pump beam present. Saturated absorption spectra of Na are shown in figure 4.8. The two maxima are resulting from situation 1, while the so-called cross-over at 0.86 GHz is due to situation 2.

In case of the 860 MHz EOM one has to lock the laser to the left flank of the cross-over around 860 MHz to assure that the -1 order can be used for cooling and trapping. Because for the 1720 MHz EOM, one uses the main frequency for pumping locking on the left flank of the first maximum leads automatically to the desired red-detuning.

The signal from the lock-in amplifier is sent to a PID (Proportional, Integral and Derivative) control (home made) the output of which is sent to the dye laser as feedback. By this, stability over several hours has been achieved, i.e., the laser frequency staying within the trapping frequency range of the MOT. At best a frequency stability of 2 MHz was reached.
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Figure 4.8: Saturation absorption spectroscopy spectra. Panel (a) shows the pure absorption signal of the probe beam as measured by the photodiode without modulation. The two maxima and the cross-over minimum are superimposed on broad absorption profiles centered around $0 \left(3s^2S_{1/2}(F=2) - 3p^2P_{3/2}\right)$ and $1770\text{ MHz} \left(3s^2S_{1/2}(F=1) - 3p^2P_{3/2}\right)$. Panel (b) shows the absorption signal with a chopped pump beam after the lock-in amplifier. To lock the laser on the desired cooling and trapping frequency either the left flank of the cross-over minimum (1, 860 MHz EOM) or the left flank of the first maximum (2, 1720 MHz EOM) is used.

4.2.5 Magneto-Optical Trap

The beam from the dye laser passes the EOM and couples into a fibre (see figure 4.7). Before the coupling into the fibre a small fraction of the laser beam is split off to an optical spectrum analyzer to monitor the intensities of the side-bands created by the EOM (not shown). The fibre (Point Source, 10 m long, 50% transmission) transports the laser beam to the MOT setup. The laser setup is placed in a separate room to shield it for dust, temperature variations, air draft and mechanical vibrations that could lead to laser instabilities.

On the optical table surrounding the vacuum chamber of the MOT the laser beam is coupled out of the fibre and passes through two acousto-optical modulators (AOM, both Isomet 1205C-2). The basic principle of the AOM is Bragg scattering of light from an acoustic wave propagating through an optically non-linear crystal. By rotating the AOM with respect to the incident laser beam the angle of incidence is varied and 90% transmission into the first order scattered beam can be obtained. The first AOM (AOM1) is used to switch off the laser beam when the ion pulse passes the MOT cloud, thereby assuring that all the sodium atoms are in the ground state, and only collisions with ground state atoms occur. If the light is off for a
Figure 4.9: Schematics of the arrangement of the two AOM’s. Part (a) shows the splitting of the primary laser beam into beams for cooling and trapping (MOT), longitudinal cooling (OVEN) and additional excitation (not used in this thesis). In (b) the typical detunings of the different beams are shown. The timing scheme of both AOM’s is depicted in (c), when AOM1 is switched off then AOM2 is turned on to reduce the intensity in the oven beam.

short enough period (20-40 µs), the MOT density is barely affected.

The +1 order of AOM1 is used as the MOT cooling and trapping beam. In a beam expander, the beam is expanded to a diameter of 2 cm. By two beamsplitters this beam is split into three equally strong trapping beams. Each trapping beam is guided through a quarter waveplate to produce the desired circular polarization. By retro-reflecting the three orthogonal trapping beams into themselves the six beams needed for a MOT are created.

The anti-Helmholz magnetic field is obtained by two parallel watercooled coils in which current flows in opposite directions. Each coil consists of 20 windings, and is provided with a current of 80 A, resulting in magnetic field gradients of ~30 Gauss/cm. Also installed are steering coils (two pairs of Helmholtz coils, ~30 windings, ~2 A) which produce a small additional homogeneous magnetic field. With these small fields the position of the zero point in the anti-Helmholz magnetic field can be moved and along with it, the MOT cloud. The steering coils can move the MOT’s y and z position over a range of ~2 mm.

The Na is evaporated from an oven containing several grams of Na. Heating takes place at rather low temperatures ($T = 130 – 150^\circ$C). The capillary of the oven exit has a diameter of 1 mm and a length of 10 mm. The oven is basically a copy of the one used in the ALCATRAZ experiment (see figure 3.9 of ref. [157]). Typically the MOT-cloud consists of a few $10^6$ cold Na atoms in a volume of 1-5 mm$^3$. In figure 4.10 a picture of the MOT could is shown.

A way to increase the number of atoms trapped in a MOT is to use an additional laser beam directed to the oven to provide longitudinal cooling (see for example [132]). This laser light is further red-detuned than that of the trapping beams. Therefore, the light decelerates atoms at higher velocities. This “pre-cooling” increases the number of atoms that can be trapped in the MOT by up to a factor of 5 [158].

The pre-cooling beam, coined “oven beam”, is provided by the second AOM (AOM2).
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The zeroth order of AOM1 is sent to AOM2 to create this oven beam (in zeroth order). The AOM arrangement is shown in figure 4.9. When AOM1 is switched off all intensity is in the zeroth order (oven beam, cf. figure 4.9). Although the oven beam is far red-detuned and only partially overlaps the MOT-cloud it might excite the atoms. Therefore also AOM2 is switched during the off time of AOM1, such that most of the intensity after AOM2 goes into the first order, not entering the MOT chamber.

4.2.6 Temperature

One reason to use laser cooling and trapping to provide a cold target for recoil-ion spectroscopy, is that the temperature is much lower than the one obtainable by the supersonic-expansion method. To determine the temperature of the atoms in the MOT-cloud, so-called “release-and-recapture” (R&R) measurements were performed [148]. In this method the temperature of the cold atoms, or more precisely their velocity, is determined by measuring the expansion of the MOT-cloud as a function of the time after switching off the trapping light.

Assuming an initial Maxwell-Boltzmann velocity distribution and a ballistic expansion after the trapping light is switched off, the fraction of atoms that is recaptured immediately after the trapping light is switched on again is given by:

$$f = \text{erf} \left( \sqrt{\frac{m}{2k_B T \Delta t}} \frac{r}{\Delta t} \right) - \frac{2}{\sqrt{\pi}} \left( \sqrt{\frac{m}{2k_B T \Delta t}} \frac{r}{\Delta t} \right) \exp \left[ - \frac{m}{2k_B T} \left( \frac{r}{\Delta t} \right)^2 \right],$$

(4.1)

in which $m$ is the mass of the atom, $\Delta t$ is the off time, $r$ is the so-called capture radius, $k_B$ is
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Figure 4.11: The depletion of the MOT-cloud after switching off the cooling and trapping light, as measured with the release-and-recapture method.

the Boltzmann constant, and $T$ is the temperature (see e.g. [159]). The measured fluorescence as function of the off-time is fitted with equation 4.1 to obtain the temperature.

To switch the trapping laser AOM1 is used. The on-off switching sequence is steered by a Labview program. The off-period is changed while the on-period is kept constant and long enough to recreate the initial MOT conditions. A CCD (charge coupled device) camera triggered every time AOM1 is switched on monitors the fluorescence of the remaining atoms. The exposure time of the camera is 100 $\mu$s, which is short compared to the retrapping time, but long enough to obtain a reasonable fluorescence signal. A typical dataset is shown in figure 4.11. From this figure it is seen that the “half-life” of the MOT-cloud is approximately 1.5 ms.

During this thesis research period R&R measurements have been performed twice. First a temperature of 380$\pm$122 $\mu$K was found [159], but recently a lower value of 176$\pm$68 $\mu$K was measured [160]. At the time of the first R&R measurements the laser frequency was not externally stabilized, which might have led to a less well-defined detuning. During the second measurement series the laser frequency was actively locked, using saturation absorption spectroscopy, and the frequency detuning was set such that the fluorescence was maximal. Note that the Doppler limit for Na is 240$\mu$K, which is a typical temperature in this kind of MOT setups. The measured temperature corresponds to a momentum spread of less than 0.01 a.u., which is equivalent to 0.5 m/s.

4.2.7 Vacuum System

Ultrahigh vacuum conditions are reached by a 240 l/s ion getter pump and a titanium sublimation pump. Without heating the Na oven a background pressure of $1-2\times10^{-10}$ mbar is obtained. In operational mode when the oven is heated to 130-150°C a Na vapor pressure in the $0.5-1\times10^{-9}$ mbar range is reached. These ultra-high vacuum conditions assure that the
4.3 Recoil-ion Momentum Spectroscopy

The ultracold Na atoms in the MOT serve as target for recoil-ion momentum spectroscopy investigations on electron dynamics occurring in ion-atom interactions. In this section the other parts of the experimental setup are described. After a brief introduction of the ion source, our recoil-ion momentum spectrometer is discussed, including the measurement principles and data analysis. Finally the experimental method used to measure charge transfer from excited Na is presented.

4.3.1 Ion source and beam transport

The projectile ions are provided by a home-built 14 GHz CAPRICE-type Electron Cyclotron Resonance Ion Source (ECRIS) [161]. An ECRIS is basically a magnetic bottle which confines a dual temperature plasma, containing hot electrons and cold ions. Multiply charged ions are produced by means of sequential electron impact ionization.

In an ECRIS plasma electrons gyrate around magnetic field lines according to their cyclotron frequency \( \omega_c = \frac{eB}{m_e} \). By applying radio frequency (RF) power the electrons gain energy at magnetic fields where their cyclotron frequency \( \omega_c \) equals the RF frequency. At a magnetic field of 0.5 T \( \omega_c \) equals 14 GHz, the RF we use. A schematic of the ECRIS is shown in figure 4.12. The combined magnetic field of the permanent hexapole magnet and solenoid coils has a so-called minimum-B configuration, in which the plasma is confined. A review and historical overview on ECRIS can be found in references [162, 163].

The main parameters which have to be tuned to optimize the ion output, are the gas pressure, RF power (0.1-0.5 kW) and the current of the solenoid on the extraction side of the source (700-1000 A). The injection solenoid’s current is usually fixed at the maximum value of the current supply (1000 A). For highly charged ions, the addition of a “support” gas can strongly increase the output of higher charge states [164]. For example for the production of

![Figure 4.12: Schematic of our Electron Cyclotron Resonance Ion Source (see text).](image-url)
C$^{6+}$ and O$^{6+}$ ions, helium is used as support gas.

The ion source can be floated up to voltages of about 26 kV. This voltage defines the energy of the outgoing ions. The extraction proceeds through a movable puller lens, which can be put onto negative voltage (0 to a few kV) to improve the beam intensity, especially for low charge states or low beam energy. Nevertheless the ion yield decreases with decreasing energy. The lowest source voltage that could be used for the MOTRIMS experiments was $\sim$ 3 kV depending slightly on ionic charge states and species.

After the ions are extracted from the source they are selected by a 110° magnet according to their mass-over-charge ratio. The selected ions are focussed by means of magnetic quadrupole triplets and guided by 45° bending magnets into one of the experimental setups of the Atomic Physics group. Typical beamline vacuum is $\lesssim 10^{-7}$ mbar, which is low enough to prevent significant losses due to charge changing collisions with background gas. After the last bending magnet in front of the MOTRIMS experiment the ions are focussed by a magnetic quadrupole doublet, after which they pass through a series of four diaphragms. The first diaphragm with a diameter of 2 mm is located 81 cm from the center of the MOTRIMS chamber. Between the first and the second (1 mm diameter) diaphragm a chopper system is placed. By applying a block pulse to one of the two condenser plates of the chopper the ion beam is swept over the second diaphragm, resulting in a pulsed beam. After that the ion beam passes two more diaphragms of 1 mm diameter. The last diaphragm is positioned at 20 cm from the collision center. Taking into account the maximum opening angle of the set of diaphragms one estimates that the ion beam has a diameter of 1.8 mm by the time it reaches the MOT-cloud [159].

The pressure in the last beamline section before the MOTRIMS chamber is on the order of $5 \times 10^{-9}$ mbar. After passing the MOTRIMS chamber the ion beam is collected in a Faraday cup. Typically (continuous) ion beam currents range between 1 and 200 nA, strongly depending on the energy and charge state of the ions. In general the highest currents are obtained for high energy, low charge state ions (e.g. 25 keV/amu H$^+$) and the lowest currents used are encountered for low energy, high charge state ions (1.1 keV/amu O$^{6+}$, extraction voltage of 3 kV). For C$^{6+}$ a few experimental runs have been performed with only $\sim$0.25 nA.

4.3.2 Recoil-ion Momentum Spectrometer

A recoil-ion momentum spectrometer usually consists of two stages. In the first (extraction region) stage a weak electrostatic field is created to extract the recoil ions from the collision center. The second stage (drift region) consists of a field free region, in which the recoil ions travel to the detector. For a homogeneous extraction, first order time focussing of ions with different starting positions along the extraction field occurs at a drift distance twice that of the distance travelled in the extraction field. This is the so-called Wiley-McLaren geometry [165]. Besides time focussing also spatial focussing is of crucial importance for any RIMS experiment. Spatial focussing means that the electrostatic field geometry corrects for the target extension perpendicular to the extraction direction.

Besides on the focussing properties the resolution depends on the actual time-of-flight of the recoil ions: the longer they travel, the larger their pattern, thus the better the resolution. For a given recoil-ion this depends on the applied voltages and the length of the drift region. By lowering the voltages defining the extraction field the acceleration is smaller. However
the extraction field should be high enough such that contact potentials do not play a role.

In order to improve the resolution the setup has been modified with respect to the previous version [26, 147]. The drift region has been lengthened from 8 to 41 cm. As a consequence the Wiley-McLaren criterium is broken and additional lenses had to be placed in front of the drift tube to recover or even improve the focussing properties. By means of SIMION ion trajectory simulations [166] optimal field shapes for both spatial and time focussing have been calculated. SIMION makes use of potential arrays that define the geometry and potentials of electrodes on which the Laplace equation is solved. It turned out that for our setup one needs three lens elements in between the extraction and drift region. In figure 4.13 the electrostatic extraction geometry (cylindrically symmetric) is shown, including equipotential lines calculated by SIMION. Five voltages are applied. A positive voltage \( V_+ \) is applied to the bottom plate, while the drift tube is at negative voltage \( V_- \). Together they create an electric field which accelerates the recoil ions from the extraction region to the drift region. The three lens elements are also on negative voltages \( V_1, V_2 \) and \( V_3 \) which can be tuned independently. Figure 4.14 shows a picture of the lens system. The rings are isolated from each other and from the drift tube by ceramic spacers. The distance from the bottom plate to the collision center is 4 cm. The total length of the extraction region including the lenses is 11 cm. The
Figure 4.14: A close up of the three-ring lens system, which is attached to the drift tube using ceramic spacers.

The drift tube is 41 cm long. For detecting Na\(^+\) recoils the voltages used are \(V_+ = +2.0\) V, \(V_- = -4.9\) V, \(V_1 = -3.4\) V, \(V_2 = -3.7\) V, and \(V_3 = -0.5\) V. The extraction field is thus less than 1 V/cm. The whole spectrometer is enclosed in a grounded tube, which contains holes for access by the laser beams, the ion beam and for observation of the MOT cloud.

After the drift region the recoil ions are detected. The detection system consists of a pair of chevron-stacked multichannel plates (MCP) in conjunction with a 2D delay line detector (DLD40). In short the MCPs amplify the recoil signal, while the delay line detector registers both time and position with a resolution of 500 ps and 0.1 mm, respectively. The complete detection system, discussed below, including the data analysis software has been developed by Roentdek [167]. In order to avoid penetration of the MCP high voltage into the drift region, a superfine electroformed Cu mesh with a 12.5 \(\mu m\) wire diameter and 88% transparency is put at the end of the drift tube in front of the MCP.

The delay line detector registers both the position and the arrival time of the recoil ions. A delay line anode makes use of the fact that the electron cloud hitting the wire, needs a measurable, finite time to reach the ends of that wire. The signals from both ends of the wire are supplied to a time-to-digital convertor (TDC) to register and digitize the arrival times. The sum of the times gives the duration to propagate along the entire wire (=constant). From the difference of the two times one can determine the position where the electron cloud has hit the wire. Two dimensional positioning is achieved by wrapping two wires perpendicular to each other and by timing the four wire ends with a one-start fourfold stop TDC (HM1).

From the propagation speed of the signal along the delay line wires, 0.71 mm/ns for the DLD40, the correspondence between position and time in the 2D position image is about 1.42 ns/mm. The TDC is used in its highest resolution mode, in which 1 channel corresponds to 133 ps. This means that for the raw data the conversion from channels to mm is given by 11 ch/mm. The quoted resolution of 0.1 mm corresponds to 1 channel. The total time range of the TDC in the high resolution mode is 2.18 \(\mu s\).

The timing scheme of the TDC used is slightly different from earlier measurements [147]. Instead of using a delayed signal from the ion beam chopper to start the TDC, one of the
detector signals ($X_2$) is used as a common start (see figure 4.15). The delayed signal from the ion beam chopper signal is used as a stop. By using cable delays the other detector stops ($X_1, Y_1, Y_2$) will always arrive later than the $X_2$ start. In this arrangement $X_1$ is directly linked to the $X$ position and $X_2$ is the inverted time-of-flight: $2.18 \, \mu s$ - time-of-flight. For the $Y$ position, $Y = Y_1 - Y_2$ still holds. With this scheme the position resolution is independent of the time-of-flight and moreover the TDC is only started if an event is detected. The latter has improved the event rate quite drastically since the TDC gets no longer blocked or occupied by starts which are not followed by stops in all four channels.

The TDC is connected to a PCI I/O card and all detected information is stored event by event in a listmode file. A C++ based program (CoboldPC: Computer Based On-line and Off-line Listmode Dataanalyzer [167]) is used to handle and analyze the listmode data. One can access this list either during data acquisition (on-line mode) or after the experiment (off-line mode).

### 4.3.3 Switching the magnetic field

A disadvantage of using a MOT to provide a cold target for recoil-ion momentum spectroscopy is the presence of the inhomogeneous magnetic field of the MOT. The slow recoil ions cross these field lines. This disturbs the recoil image on the detector. For longitudinal extraction the longitudinal momentum component is not affected, because the extraction is along the field lines, but both transverse momentum components are disturbed [25, 168]. For transverse extraction the longitudinal momentum and one of the transverse momenta are...
affected [146, 152]. In part one can correct for these effects.

In order to circumvent this problem completely a current supply has been designed and built which is able to switch the MOT coils on and off such that during the measurement cycles the magnetic field is not present. By its design, it is possible to switch off a current of 80 A within 6.7 $\mu$s. The “Magnetic Field Switcher” is able to run at frequencies up to 8 kHz. This fast switching could not be used. Switching the high current in the MOT coils at several kHz causes eddy currents in the stainless steel vacuum chamber, thereby heating the vacuum chamber. The heating resulted in an increase of the vacuum pressure by two orders of magnitude.

It was realized that a much lower frequency could be used because of optical molasses cooling by the laser beams. Fig. 4.16 shows two R&R measurements, one in which the laser beams are switched off and one in which the magnetic field is switched off but the laser beams are kept on. The beneficial effect of the optical molasses is obvious. After 20 ms 80% of the original MOT-cloud intensity is still left.

In order to measure recoils only if the magnetic field is switched off a start signal is sent to the TDC only if the current of the “Magnetic Field Switcher” is turned off. When the target density drops, the magnetic field is switched on again to recover the MOT. The switching frequency is 100-150 Hz, the chopping frequency (i.e. the repetition rate of the measurement) is 6.5-7 kHz (see figure 4.17). At frequencies well below 1 kHz, no heating due to eddy currents was observed.

### 4.3.4 Data analysis

For clarity a schematic of the MOTRIMS setup is shown in figure 4.18, in which also the momentum axes are indicated. The longitudinal momentum axis is defined by the ion beam,
the extraction towards the detector is along the Z-axis. Since the target is unpolarized, the collision system possesses cylindrical symmetry around the beam axis. Therefore, only the momenta in longitudinal ($p_{\text{long}}$) and in transverse ($p_{\text{trans}}$) direction to the ion beam are relevant.

A 2D detector image is depicted in figure 4.19, which shows Na$^+$ recoils resulting from O$^{6+}$+Na collisions. Perpendicular to the $X$-direction (the ion beam direction) lines are observed. The lines arise from capture into different $n$-shells of O$^{5+}$. From the projection of the 2D spectrum on the $X$-axis one can obtain the relative intensities of these lines, as shown in figure 4.20 (the $X$-position is converted into Q-value).

To translate the measured $XY$-distribution into the physical quantities of interest the following transformations have to be made. The longitudinal direction, i.e. $X$-direction, is perpendicular to the extraction field, which means that no external force is acting in this direction. Therefore the relation between the longitudinal momentum $p_{\text{long}}$ and the $X$-position of the recoil-ion on the detector is given by

$$p_{\text{long}} = M \frac{X}{T}, \quad (4.2)$$

in which $X$ is the $X$-position of the recoil-ion on the detector, $M$ is the mass of the recoil-ion, $T$ is the time-of-flight. The spread in $T$ for a given recoil charge state is usually small compared to the total time-of-flight. For instance, in the normal extraction setting $T = 63$ $\mu$s for Na$^+$ recoils, while the FWHM of the Na$^+$ time-of-flight peak is smaller than 0.5 $\mu$s. Therefore $T$ can be taken to be a constant. This implies that the longitudinal momentum is simply proportional to the $X$-position on the detector [112], i.e.,

$$p_{\text{long}} \approx \alpha X, \quad (4.3)$$

in which constant $\alpha$ links a position $X$ in channel numbers to a momentum in atomic units. Constant $\alpha$ takes different values for different extraction potentials and recoil charge states, because it scales with the time-of-flight. Finally, the Q-value spectra can be obtained from
rewriting equation 3.10,

\[ Q = v_p (p_{long} + \frac{1}{2} r v_p). \] (4.4)

The transverse momentum, \( p_{trans} \), is a 2 dimensional vector, with components \( p_{trans,y} \) and \( p_{trans,z} \). Its magnitude is given by

\[ p_{trans} = \sqrt{p_{trans,y}^2 + p_{trans,z}^2}. \] (4.5)

Experimentally the transverse momentum components are obtained in quite different ways. Because the \( Y \)-direction is perpendicular to the extraction direction the same argumentation can be made as for the longitudinal momentum,

\[ p_{trans,y} \approx \alpha Y. \] (4.6)

Note that \( \alpha \) is exactly the same as in case of \( p_{long} \) (equation 4.3).

The momentum component \( p_{trans,z} \) is less trivial to obtain, because the recoils are accelerated along the \( Z \)-axis. It is connected to the time-of-flight via the following equation,

\[ T = -v_{trans,z} + \frac{\sqrt{v_{trans,z}^2 + 2 z_e a}}{a} + \frac{z_d}{\sqrt{v_{trans,z}^2 + 2 z_e a}}, \] (4.7)
in which \( v_{\text{trans},z} = p_{\text{trans},z}/M \), \( z_d \) the distance from the collision center to the entrance of the drift region, \( z_d \) the length of the drift region. The acceleration \( a \) is given by \( a = rE_z/M \), in which \( E_z \) is the electric field. The derivation of equation 4.7 assumes a homogeneous electric field. For an inhomogeneous field it can still be applied by taking \( E_z \) as the “effective” electric field experienced by the recoil ions. The exact inversion of equation 4.7, needed to calculate \( p_{\text{trans},z} \), is rather complicated [111]. One can circumvent this problem by again using the fact that differences in time-of-flight between the recoil ions are small as compared to the average time-of-flight \( T_0 = T(v_{\text{trans},z} = 0) \). Expanding equation 4.7 around \( T_0 \) leads to the relation

\[
\Delta t \equiv T - T_0 \approx -\frac{v_{\text{trans},z}}{a},
\]

in which \( \Delta t \) can take positive and negative values depending on the sign of \( v_{\text{trans},z} \). A recoil momentum directed towards the detector has a positive \( v_{\text{trans},z} \) and arrives earlier at the detector than a recoil-ion with zero momentum in the \( Z \)-direction, thus \( \Delta t < 0 \).

Most importantly, only the deviation from \( T_0 \) is needed to obtain the transverse momentum in the \( Z \)-direction, which can be represented as

\[
p_{\text{trans},z} \approx -\beta \Delta t,
\]
Experimental setup

Figure 4.20: $Q$-value spectrum of Na$^+$ recoils, resulting from 2.25 keV/amu O$^{6+}$+Na collisions, obtained from projecting the XY-distribution shown in figure 4.19 onto the X-axis. The measured X-position is related to the Q-value via equations 4.2 and 4.4.

in which $\beta$ is a constant depending on the extraction potential and geometry. This approximation is valid if the initial kinetic energy of the recoil-ion is much smaller than the energy gained in the extraction field, i.e. $p_{trans,z}^2/2M \ll rE_z e$. This condition is fulfilled even for the lowest extraction fields used. For example, a field strength of 0.5 V/cm over 7 cm leads to an extraction energy of 3.5 eV. Typical transverse momenta are smaller than 1 a.u. which corresponds to a kinetic energy of less than 1 meV, which is thus more than a factor 1000 smaller than the energy gained in the extraction field. For recoil momenta much larger than 1 a.u., one also uses higher extraction fields to collect all or almost all recoil ions on the detector.

For the Na$^+$ recoil spectra typically a resolution of 0.1 a.u. (corresponding to 5 m/s) was achieved in the longitudinal, $p_{long}$ and in one of the transverse directions, $p_{trans,y}$. This resolution is obtained with the voltage settings listed above, and allows to resolve the main $n$-shell capture channels. The extraction is efficient enough to extract all recoil ions towards the detector. A longitudinal momentum spectrum of Na$^+$ recoils resulting from He$^{2+}$+Na collisions is shown in figure 4.21. The resolution is defined by the width of the capture peaks, which are assumed to be Gaussian shaped. From this spectrum it is also seen that the signal-to-noise ratio is so high that even the weak $n = 2$ capture channel can be observed.

For higher charge state recoils the resolution is somewhat worse. For a given extraction potential the time-of-flight is inversely proportional to the square root of the charge state, which means that the momentum image on the detector is compressed for higher charged recoils. Also higher extraction potentials are usually needed to collect all recoils, because they have larger transverse momenta as compared to the Na$^+$ recoils. This shortens the time-of-flight somewhat and therefore the resolution is reduced.

Recently the resolution of the apparatus has been improved to 0.07 a.u. (3 m/s) by lower-
4.3 Recoil-ion Momentum Spectroscopy

Figure 4.21: A longitudinal momentum spectrum of 4 keV/amu He$^{2+}+Na(3s)$. Capture into $n=3$ and $n=4$ can be clearly separated. The inset shows the same spectrum on a log scale, from which capture into $n=2$ can be seen. The resolution obtained is 0.1 a.u.

By further reducing the extraction voltages to half of their values, the recoil patterns start to become distorted. This might be due to contact potentials and stray fields. Figure 4.22 compares a 0.1 and 0.07 a.u. resolution spectrum of Na$^+$ recoil from 6 keV/amu $^{13}$C$^6$+Na(3s) collisions. Practically a better resolution means that one can extract the probability of capture into higher $n$-shells. Note that the momentum spread of the target atoms is $\leq 0.01$ a.u. (section 4.2.6) which is still well below our resolution of 0.07 a.u.

Because of better resolution in the position than in the timing (a 30 ns ion pulse width corresponds to a momentum spread of more than 0.2 a.u.) the transverse momentum distribution is deduced from the position distribution alone. Due to the cylindrical symmetry around the beam axis the projection of the transverse momentum distribution onto the measured one ($p_{trans,y}$) is described by an Abel transformation [146]. Directly applying the inverse Abel transformation to obtain the transverse momentum distribution turns out to be quite sensitive to statistical noise. Therefore we have used a different, iterative method (see [169] and appendix B), originally developed by Vrakking [170] to extract velocity and angular distributions from two-dimensional ion/photoelectron imaging experiments.

For low count rate and noisy data, unphysical features may still show up. To overcome this an extra smoothing procedure was introduced. The raw $p_{trans,y}$ distribution was fitted by smooth functions. It has been found that spectra are well fitted by a few Gaussian distributions. The Gaussians are then used as input for the iterative method. Note that also the direct application of the inverse Abel transformation is possible, but due to its ease, the iterative method was preferred. Figure 4.23 illustrates the smoothing and transformation procedure
Figure 4.22: Comparison of longitudinal momentum spectra of Na\(^+\) recoils resulting from 6 keV/amu \(^{13}\text{C}\)\(^6\)+Na(3s). One spectrum is taken with the best resolution of 0.07 a.u. (——) and the other one with a typical resolution of 0.1 a.u. (⋯).

using the data of one-electron capture into \(n = 3\) and \(n = 4\) for 3 keV/amu He\(^2\)++Na. The \(p_{\text{trans,y}}\) raw data are smoothed using three Gaussian distribution. The iterative method is applied to both the raw data as well as the smoothed distribution. Comparing the results shows almost identical distributions for the \(n = 3\) channel, while for the \(n = 4\) channel the noise in the raw data seems to be amplified in the end result. The smoothing procedure is especially of use for low statistics data. One has to realize that this smoothing procedure reduces the resolution in \(p_{\text{trans}}\). In general not much structure was observed in the \(Y\)-distributions.

The transverse momentum is linked to the scattering angle, \(\theta\), by

\[
p_{\text{trans}} = m_p v_p \theta, \tag{4.10}
\]

where \(m_p\) and \(v_p\) are the mass and the velocity of the projectile, respectively. Equation 4.10 is valid for small angles. For one-electron capture this condition is nearly always fulfilled (\(\theta < 1\) mrad). From the transverse momentum distribution one can obtain the differential cross section (DCS), which simply means that one plots the cross section as a function of scattering angle \(\theta\). By selecting the transverse momenta within a specific longitudinal momentum bin the DCS for specific capture channels can be obtained.

In conclusion, the only two recoil momentum components needed to obtain all the information, i.e. Q-value and scattering angle, are \(p_{\text{long}}\) and \(p_{\text{trans,y}}\). As explained above these components can be determined to a very good approximation independently of the time-of-flight. From this, and from the fact that the amount of Na\(^+\) recoils is much larger than that of the other Na\(^{2+}\) recoils (see below), it was realized that Na\(^+\) recoil spectra could also be obtained with a continuous instead of a pulsed ion beam. The detector trigger is synchronized with the switching scheme of the AOM’s. Main advantage is the much higher count
4.3 Recoil-ion Momentum Spectroscopy

Figure 4.23: The $Y$-distributions of one-electron capture into $n = 3$ (a) and $n = 4$ (c) for 3 keV/amu He$^{2+}$+Na collision, showing raw data (■) and fitted distribution (—), using three Gaussian functions (···). The results of applying the inverse Abel transformation using the iterative method to obtain the transverse momentum distributions is shown in panel (b) and (d).

rate, which means that also good statistics can be obtained in the weak capture channels. The possible Na$^{2+}$,$^{3+}$,... recoils do not disturb the spectra significantly, not only because of their small cross section with respect to the Na$^+$ recoils, but also because their momenta are much higher, which means that not all of them reach the detector. Comparing Na$^+$ recoil spectra obtained from a pulsed and a continuous beam confirmed that this method could be applied.

For measuring the higher charge state recoils a pulsed ion beam is unavoidable. In this case the detector was triggered such that the time-of-flight peak of the Na$^{r+}$ recoils of interest falls inside the 2 $\mu$s time window of the TDC. A reasonable Na$^+$ spectrum can be obtained within a few minutes. Longer times up to one or two hours are needed to obtain good statistics in the weak channels.

4.3.5 Calibration of the recoil spectra

For the Na$^+$ recoils the zero-point calibration of the recoil spectra and the conversion from $XY$-channels to momenta is relatively easy because of the discrete nature of the longitudinal momenta. For one-electron capture the main capture channels are well known. From the binding energies of these, the corresponding longitudinal momenta are calculated using
Identifying two capture lines is sufficient to find the conversion from channels to momenta (i.e. constant \( \alpha \), cf. equation 4.3) and to determine the zero point. For the normal extraction settings this conversion is typically 55 ch/a.u., i.e., 1 a.u. of momentum is covered by 55 channels. A resolution of 0.1 a.u. means that the FWHM of the capture peaks covers 5 or 6 channels. For the recently applied lower extraction settings, the recoil image is enlarged on the detector and a conversion around 80 ch/a.u. was found.

For the higher \( \text{Na}^{+}\) recoil charge states, the spectra exhibit very often no sharp capture features. Here one takes advantage of the so-called “MOT-ions”, which are \( \text{Na}_2^+ \) molecules produced via associative ionization of excited Na atoms \[171, 172]\.

\[
\text{Na}^+(3p) + \text{Na}^+(3p) \rightarrow \text{Na}_2^+ + e^-.
\] (4.11)

The production of these ions depends quadratically on both the MOT density and the excited fraction \[173\]. Because these ions are produced with almost no kinetic energy, i.e. zero momentum, they can serve to mark the zero position. Of course this could also lead to an unwanted contribution in the recoil spectrum. For the \( \text{Na}^+ \) recoil spectrum this contribution is usually not significant. In case of higher charge state recoils the situation is quite different, and this part of the spectrum has to be cut out off-line to insure that the MOT-ions do not contribute to the momentum spectra.

Using the well-known \( \text{Na}^+ \) recoil spectrum it was found that the position of MOT-ions does slightly differ from the zero position. This can be explained by imperfections in the extraction field or stray magnetic fields leading to a displacement perpendicular to the extraction direction. Such a displacement will depend on the mass-over-charge ratio. Assuming that the dependence is similar to that of the time-of-flight, namely proportional to the square root of the mass, the difference in zero-position of the \( \text{Na}^+ \) \((M = 23)\) recoils and the MOT-ions \((M = 46)\) can be used to determine the “real” zero-point of the other recoil charge states. Any recoil measurement of higher charge state recoils has to be accompanied by a measurement of the \( \text{Na}^+ \) recoils in the same experimental settings. Also the conversion of channels to momenta obtained from \( \text{Na}^+ \) recoils can be used for the higher charge state recoil spectra, realizing that this conversion depends linearly on the time-of-flight (see equation 4.2 and 4.3). Thus the conversion factor scales with the square root of the recoil-ion’s charge state.

### 4.3.6 Extracting partial cross sections

From the \( \text{Na}^+ \) Q-value spectra the relative cross sections of the different processes can be extracted. The relative contributions of capture into the different \( n \)-shells are obtained by fitting the spectra, assuming Gaussian peak shapes. This fitting is usually only possible for capture peaks that are sufficiently resolved. Therefore separate partial cross sections are only determined up to a certain \( n \)-shell. For higher \( n \)-shells one can obtain only summed cross sections.

\( \text{Na}^+ \) recoils can also be created by single ionization. This process leads to values of \( Q \) larger than the ionization potential \( I = 5.14 \) eV. It can be shown that ionization events cannot appear at the capture side of the recoil spectrum and for those recoils

\[
P_{\text{long}} > \frac{I}{v_p} - \frac{1}{2}v_p^2.
\] (4.12)
Therefore $Q = I$ marks the Q-value boundary between capture and ionization contributions [115, 174, 175]. Taking the experimental resolution into account and assuming a continuous decrease for $Q \rightarrow I$ ($n \rightarrow \infty$) the relative contributions of capture into high-$n$ shells and ionization can be extracted. Furthermore, one can show that the Q-value of the ionization part of the spectrum is directly connected to the energy of the emitted electron in the projectile frame (see appendix C).

If $n_{\text{max}}$ is the highest $n$-shell for which separate cross sections can be obtained by the fitting procedure, then the sum contribution of capture into $n > n_{\text{max}}$, i.e.,

$$\sigma_{n>n_{\text{max}}}^{\text{rel}} \equiv \sum_{n=n_{\text{max}}+1}^{\infty} \sigma_n^{\text{rel}}, \quad (4.13)$$

is determined as follows:

$$\sigma_{n>n_{\text{max}}}^{\text{rel}} = \sigma_{\text{tot}}^{\text{rel}} - \sum_{n=1}^{n_{\text{max}}} \sigma_n^{\text{rel}} - \sigma_{\text{ion}}^{\text{rel}}, \quad (4.14)$$

in which $\sigma_{\text{tot}}^{\text{rel}}$ is the total recoil spectrum, $\sum_{n=1}^{n_{\text{max}}} \sigma_n^{\text{rel}}$ the contribution of the capture peaks that can be fitted individually and $\sigma_{\text{ion}}^{\text{rel}}$ is the ionization contribution.

The separation of capture and ionization processes is not only possible for Na$^+$ recoil spectra, but also for higher recoil charge states. For example, in Na$^{2+}$ recoil spectra transfer ionization can be separated from two electron capture. Appendix C presents more details on extracting the ionization part of the recoil spectrum.

### 4.3.7 Absolute cross sections

The relative cross sections for electron capture and ionization are obtained from the Q-value spectra. In order to compare directly with theory and other experiments, one would like to put the results on an absolute scale. It has turned out that it is not readily feasible to achieve an absolute calibration better than 50% because we have to control many if not all experimental parameters quite precisely. These parameters are the number of incoming ions, the ion beam profile, the target density and the overlap between the ion beam and the target cloud. The number of ions is easily accessible by measuring the ion beam current in a Faraday cup. The beam profile is difficult to determine. It can be estimated from the positions and sizes of the diaphragms. For the target density both the number of trapped atoms and the target profile is needed. The latter can be obtained from the CCD images which are taken during the experiments, but the number of trapped atoms cannot be obtained very accurately. Secondly, but most importantly, the exact knowledge of the overlap between the ion beam and the target cloud is very difficult to obtain (see also discussion in [152]). Therefore, our data are normalized to absolute cross sections available in the literature. Knowing an absolute one-electron capture cross section ($\sigma_{\text{cap}}$) our relative partial capture cross sections ($\sigma_n^{\text{rel}}$) can be made absolute by normalizing the sum of all capture channels to $\sigma_{\text{cap}}$, i.e.,

$$\sigma_n = \frac{\sigma_n^{\text{rel}}}{\sum_n \sigma_n^{\text{rel}}} \sigma_{\text{cap}}, \quad (4.15)$$
Experimental setup

4.3.8 Time-of-flight measurements

Because the time-of-flight window of the TDC is 2.2 $\mu$s whereas ions with different charge state arrive typically more than 5 $\mu$s apart, only the momenta of one recoil charge state can be measured at a time. In order to compare the relative cross sections for different recoil charge states time-of-flight measurements were performed. One of the detector’s delay line signals is used to stop a TAC (Time-to-Amplitude Convertor), started by the chopper signal. The TAC can handle times up to 300 $\mu$s. To have full detection efficiency for all recoil charge states extraction fields of 2 to 20 V/cm were used.

Besides the Na$^{+}$ peaks also contributions from the background appear in the time-of-flight spectra, mostly from H$_2$O$^+$, H$_2^+$ and H$. Figure 4.24 shows a typical time-of-flight spectrum resulting from 6 keV/amu He$^{2+}$+Na collisions. Note that the presence of background recoils is enhanced because they are formed along the whole ion beam path “seen” by the detector ($\sim$ 5 cm), while the Na target has a diameter $\leq$ 1 mm only.

By integrating the Na$^{+}$ peaks one obtains relative cross sections for one- or more-electron removal. Typically the amount of Na$^{2+}$ is only a few percent of Na$^+$. Knowing the Na$^+$ production cross section also for multi-electron removal processes the absolute cross section can be obtained.

\[ \sigma_{\text{ion}} = \frac{\sigma_{\text{ion}}^{\text{rel}}}{\sum_n \sigma_{\text{cap}}^{\text{rel}}} \]
4.3 Recoil-ion Momentum Spectroscopy

4.3.9 Collisions with excited Na

As already pointed out the MOTRIMS experiments are very suitable to study charge transfer from excited atoms (see section 3.3). Here the experimental method used to study collisions with Na atoms in the excited $3p$ state is described.

For the experiments with ground state Na the AOM’s are periodically switched, such that during the collisions no laser light is present and all Na atoms have decayed to the ground state. Without this switching the recoil spectra contain contributions from collisions on both ground state, Na$(3s)$, and excited state, Na$^*(3p)$. In order to extract the contribution from the latter, recoil spectra with and without switching the AOM’s have to be subtracted in an appropriate manner.

We define the cross sections for the production of Na$^+$ recoils from Na in the ground state to be $\sigma(3s)$ and with Na in the excited state $\sigma(3p)$. If the laser beams are off the recoil signal $S_{\text{off}}$ is given by:

$$ S_{\text{off}} = A\sigma(3s), \quad (4.17) $$

in which $A$ is a constant depending on various experimental parameters. If the laser beams are on, the recoil signal $S_{\text{on}}$ is given by:

$$ S_{\text{on}} = B[(1-f)\sigma(3s) + f\sigma(3p)] = B(1-f)[\sigma(3s) + \frac{f}{1-f}\sigma(3p)], \quad (4.18) $$

In case all experimental conditions are the same when measuring $S_{\text{off}}$ and $S_{\text{on}}$, then $A = B$. Parameter $f$ is the excited fraction, i.e. the fraction of Na$^*(3p)$ atoms.

Two approaches to extract the contribution from Na$^*(3p)$ can be followed. The simplest way is to normalize both $S_{\text{off}}$ and $S_{\text{on}}$ spectra on a capture peak resulting from the ground state and well resolved from any excited Na contribution. Calling the cross section of this normalization channel $\sigma(3s)_{\text{norm}}$, then the normalized recoil signals $S_{\text{off}}^{\text{norm}}$ and $S_{\text{on}}^{\text{norm}}$ are given by:

$$ S_{\text{off}}^{\text{norm}} = \frac{S_{\text{off}}}{A\sigma(3s)_{\text{norm}}} = \frac{\sigma(3s)}{\sigma(3s)_{\text{norm}}}, \quad (4.19) $$

and

$$ S_{\text{on}}^{\text{norm}} = \frac{S_{\text{on}}}{B(1-f)\sigma(3s)_{\text{norm}}} = \frac{\sigma(3s)_{\text{norm}} + f}{\sigma(3p)_{\text{norm}}}, \quad (4.20) $$

from which it is easily seen that by subtracting these normalized signals the contribution from the ground state is removed:

$$ S^{3p} = S_{\text{on}}^{\text{norm}} - S_{\text{off}}^{\text{norm}} = \frac{f}{1-f}\sigma(3p)_{\text{norm}} = C\sigma(3p), \quad (4.21) $$

whereby $C$ is a constant. From the resulting spectrum $S^{3p}$ the relative partial cross section can be obtained. From this procedure only relative partial cross sections of capture and ionization from the excited state can be obtained. To obtain absolute cross sections one needs to know the excited fraction $f$.

A second approach is based on having equal experimental conditions, i.e. $A = B$. These conditions can be met in measurements of a few minutes and by actively monitoring the
ion beam current. By measuring alternatingly spectra with and without excited atoms, the stability of the experimental conditions can be checked off-line. Explicitly, for $A = B$ one can write down equation 4.17 and 4.18 in the following forms:

$$S^{\text{off}} = A \sigma(3s)$$  \hspace{1cm} (4.22)

$$S^{\text{on}} = A[(1-f)\sigma(3s) + f\sigma(3p)].$$  \hspace{1cm} (4.23)

To subtract these signals directly one needs first to determine the excited fraction $f$. This can be done by looking at the decrease of the normalization peak which is solely due to Na(3s) [143, 176]. In comparing $S^{\text{off}}$ and $S^{\text{on}}$ this peak should be lowered by a factor $(1-f)$.

After one has found $f$ the two signals have to be subtracted in the following way:

$$S^{\text{on}} - (1-f)S^{\text{off}} = Af\sigma(3p).$$  \hspace{1cm} (4.24)

The ratio between one-electron removal from Na(3s) and Na*(3p) is now given by:

$$\sigma(3p)/\sigma(3s) = \frac{S^{\text{on}} - (1-f)S^{\text{off}}}{fS^{\text{off}}}.$$  \hspace{1cm} (4.25)

Advantage of this method is the determination of the ratio of Na+ recoil production from excited and ground state Na. If absolute cross sections for Na(3s) are available, also processes connected to the excited state can be put on an absolute scale. Typical excited fractions were about 15-20%, which is clearly lower than the 30% found in previous TES [177] and PES [88] measurements. A possible explanation is the direct use of the near-resonant laser light applied for cooling and trapping, and not resonant light.

A similar scheme is used by the Kansas group in their MOTRIMS experiment [27, 144, 168]. However, in their case the AOM switching status is added directly to the data acquisition, labelling each recoil event whether to originate from an on or off period. This could not be applied here because the used TDC does not allow to integrate this information into the data collection.

As a final remark on the properties of the excited atoms, one assumes that the sample of Na*(3p) atoms in a MOT is unpolarized. Because the cold atoms are excited by three orthogonal pairs of counterpropagating laser beams there is no preferred alignment or orientation direction with respect to any quantization axis. One can consider equal populations for the $m$-substates or equivalently for the atomic orbitals $\sigma, \pi^+$ and $\pi^-$ of the Na*(3p) state. Thus in comparing our $\sigma(3p)$ cross sections with previously reported cross sections of capture from aligned or oriented Na*(3p), one has to take the average over the $m$-substate or atomic orbital specific cross sections, i.e.,

$$\sigma(3p)_{\text{MOT}} = \frac{1}{3}[\sigma(3p_0) + \sigma(3p_{+1}) + \sigma(3p_{-1})]$$  \hspace{1cm} (4.26)

$$= \frac{1}{3} [\sigma(3p\sigma) + \sigma(3p\pi^+) + \sigma(3p\pi^-)].$$  \hspace{1cm} (4.27)

Further remarks on the alignment and orientation description of Na*(3p) and the polarization properties in the MOT are given in appendix D.